NONDESTRUCTIVE ANALYSIS OF IRRADIATED REACTOR FUEL BY GAMMA RAY SPECTROSCOPY

by

THOMAS J. KELLEY

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ABSTRACT

The feasibility of performing nondestructive analysis of spent power reactor fuel by means of gamma ray spectroscopy was established. Dose rates to be expected from spent power reactor fuel pins offered by the AEC for study were calculated. Parallel calculations were performed for MITR fuel elements and the results compared with measured dose rates of the MITR fuel elements. Gamma ray spectra were obtained from MITR fuel elements and used to estimate the expected gamma ray spectra for the spent pins. Calculations were performed to determine the number of Pu-239 fissions which had occurred in the spent fuel pins and the results used to calculate the expected change in the spectra due to the presence of Pu-239 fission product gamma rays. Recommendations are made to enhance the ability of M.I.T. researchers to perform the subject experiments on actual spent power reactor fuel.

Thesis Supervisor: Michael J. Driscoll
Title: Associate Professor of Nuclear Engineering

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I: INTRODUCTION

The possibility of utilizing gamma ray spectroscopy for determination of fission product content for use in ascertaining the irradiation history of spent fuel has been studied by many investigators (1-4). Sovka (5) utilized the excellent energy resolution of Ge(Li) detectors to develop a method for determining spatial distribution of absolute neutron flux, neutron exposure, total U-235 burnup and irradiation time of the fuel. This method was unique in that it did not require absolute calibration of the detecting system since it used ratios of fission product activities. However, because it does use ratios of fission product activities, more than one fission product's gamma ray(s) must be resolved. This work was performed using the spectra obtained from spent MITR fuel. Since MITR fuel has a pre-irradiation enrichment of 93%, it differs considerably from that of the more widely used and economically more important PWR or BWR power reactor fuel which usually has an enrichment of 2-4%. Because of the importance of accurate knowledge of the present status and past history of partially burned or spent power reactor fuel from the viewpoint of fuel cycle economics, reactor core physics, and most recently, safeguards, it is quite important that advanced spectrometry methods be extended to these fuel types. This extension is in part, one of the aims of the M.I.T. Reactor Physics Project which is sponsored by the AEC and under

whose aegis the present work was performed. For example, the power reactor fuel with its high percentage of U-238 would have a significant number of Pu-239 fissions, and thus the spectra from spent power fuel would be altered because of the difference in the fission product yields for U-235 and Pu-239. Self-shielding effects upon the emitted spectra would also obviously differ. The purpose of this study was therefore to determine the feasibility of performing gamma ray spectroscopy on spent power reactor fuel.

In order to establish feasibility, gamma ray spectra were obtained from spent MITR fuel to measure the energy resolution of the detecting system, to obtain data from which to estimate the relative intensities of gamma rays present in power reactor fuel spectra, and to determine modifications to the scanning system necessary for the scanning of power reactor fuel by subsequent investigators.

Calculations of the dose rate to be expected from the spent fuel offered by the AEC for study were made in order to develop safe handling procedures for the highly radioactive fuel, which was in turn necessary in order to obtain AEC licensing for the subject experiments. Parallel calculations were performed on spent MITR fuel and comparisons made with observed dose rates in order to check the fidelity of the calculations and to normalize the predicted results to physically measured quantities. Finally, calculations of the relative gamma ray intensities present in the power reactor fuel spec-

tra after a 2.75 year cooling period were made and their results used to determine the suitability of the proposed fuel for analysis. Each of the above aspects are reported upon in detail in the three chapters which follow.

II: DOSE RATE CALCULATION FOR SPENT FUEL

2.1 Introduction

Prior to obtaining spent power reactor fuel it was necessary to develop handling procedures and to secure AEC licensing approval, which necessitated estimation of the dose rate expected from the spent fuel. Two fuel pins which had been tested in the Dresden power reactor were made available by the AEC for the studies. Some of the known and calculated information (6) on the fuel pins, numbered A4 and A28 in the Dresden core loading is listed in Tables 1 and 2.

2.2 Background Information

Many studies (7-11) have been published on the activity of spent fuel ranging from experimental studies based on calorimetric methods to theoretical calculations. The experimental studies present only total activities and as such are unsatisfactory for dose rate claculations, since the energy dependence of the activity must be known, because the mass absorption coefficient, the buildup factor and flux required for a given dose rate are all strongly dependent on energy. The theoretical compilations have utilized fission product data to calculate the activities for varying irradiation and decay times. Blomeke and Todd (12) have calculated the activities of specific fission products, and while this study thus gives the complete energy dependence of the spectra, the number of

TABLE 1: IRRADIATED FUEL PIN DATA

FUEL PIN	ORIG. Wt. U (gms. metal)	ORIG. Wt. U-235 (gms. metal)	ORIG. ENRICHMENT	FINAL Wt. U (gms.)	FINAL Wt. U-235 (gms.)	FINAL ENRICHMENT	FINAL Wt. Pu. (gms.)	TOTAL ENERGY MWD	MWD/T*
A4	610.10	16.84	2.76%	588.0	3.5	0.6%	4.0	15.32	22000
A28	618.64	17.07	2.76%	597.0	3.6	0.6%	4.0	17.08	23000

a) Reference (6)

*Megawatt days per metric ton (1000 kg) oxide

TABLE 2: FUEL PIN IRRADIATION HISTORY

FUEL PIN	DATE CHARGED	DATE DISCHARGED	TOTAL ENERGY MWD	DATE CHARGED	DATE DISCHARGED	TOTAL ENERGY MWD
A4	1/11/61	12/9/63	7.66	6/9/64	1/13/67	7.66
A28	6/7/61	12/9/63	8.54	6/9/64	1/13/67	8.54

fission products makes this method too cumbersome for estimating the dose rate from spent fuel. Most studies divide the energy spectrum of fission product gamma rays into two or more energy groups and sum all gamma rays whose energies fall within these groups. If a sufficient number of groups is used the calculations will account for the energy dependence mentioned previously without the necessity of accounting for each fission product's activity on an individual basis.

The study by Prawitz et al (7) was chosen to estimate the activity of the spent fuel because a sufficient number of energy groups are used and the decay of the activities is presented for cooling times up to 20 years, whereas many studies do not report the decay for times longer than one year. The study gives the activity of fission products as a function of both irradiation time and cooling time and some indication is also given of the trend for flux variations. This latter parameter is important only for fission products which have high neutron absorption cross sections. Statistically, this group of fission products represents only a small fraction of the total fission product activity. If the flux dependence must be known the comprehensive study of Blomeke and Todd (12) presents the activity of spent fuel for various irradiation and cooling times as a function of neutron flux. While this study does give the dependence of activity on flux it was felt this advantage was offset by the fewer number of energy groups

used.

2.3 Method of Calculation

In order to ensure a conservative estimate and to simplify the calculations, it was assumed the burnup occurred during a one year irradiation period, followed by a cooling time of 700 days.

The method of calculation will be illustrated for fuel pin A28 with a 4 foot water shield.

1) The 15 energy groups and their energy range are listed in columns 1 and 2 of Table 3. The effective energy of each group was assumed to be equal to the midpoint energy of each of the groups and this energy is listed in column 3 of Table 3. This energy was used for determining the values of the mass absorption coefficient, buildup factor and the flux to dose rate conversion factor to be used with the various energy groups. The release of an amount of energy equal to the effective energy was also assumed to represent one disintegration. The specific energy release rates for the various energy groups for an irradiation time of one year and 700 days cooling time are listed in column 4 of Table 3. The specific energy release rates were converted to specific activity by dividing by the effective energy for each group to convert from Mev/gFP minute to disintegrations/gFP second. This specific activity is listed in column 5 of Table 3. Specific activity was converted to Activity by multiplying by

TABLE 3: GAMMA SPECTRUM EMMITED BY FUEL

ENERGY GROUP NO.	ENERGY RANGE (MeV)	EFFECTIVE ENERGY E_γ	ENERGY gFP min.	DIS. gFP sec.	DIS. sec.
1	.05-.20	.125	6.48E11	5.18E12	1.472E12
2	.02-.40	.30	2.9E9	9.66E9	2.76E9
3	.40-.60	.50	4.23E11	8.4811	2.42E11
4	.60-.80	.70	4.41E12	6.30E12	1.792E12
5	.80-1.00	.90	2.12E10	2.36E10	6.72E9
6	1.00-1.20	1.10	6.90E10	6.28E10	1.788E10
7	1.20-1.40	1.30	3.30E9	2.54E9	7.24E8
8	1.40-1.60	1.50	1.55E11	1.03E11	2.94E10
9	1.60-1.80	1.70	1.25E10	7.35E9	2.10E9
10	1.80-2.00	1.90	6.8E9	3.58E9	1.02E8
11	2.00-2.20	2.10	5.11E11	2.44E11	6.96E10
12	2.20-2.40	2.30	1.66E10	7.22E9	2.06E9
13	2.40-2.60	2.50	-	-	-
14	2.60-2.80	2.70	-	-	-
15	2.80-3.00	2.90	-	-	-

NOTE: $EN = 10^N$

the burnup experienced by the fuel pin.

2) Once the activities of the energy groups are known, they must be converted to fluxes present at the point in question. Reference (13) contains expressions relating flux to activity, source geometry and shield thickness. For a cylindrical source of finite length the following expression is given for the geometry shown in Figure 1:

$$\phi = B \frac{S_v r^2}{2(a + Z)} \left[F(\theta, b_2) \right] \qquad (1)$$

where:

ϕ = undirected flux (γ rays/cm^2-sec)

S_v = volume source strength (γ rays/cm^3-sec)

B = buildup factor to correct for multiple scattering

r = radius of cylindrical source (cm)

a = distance from source to observer (cm)

z = effective self-absorption distance (cm)

$F(\phi, b_2) = \int_o^\theta e^{-b} \sec \theta d\theta$

$b_2 = b_1 + \mu_s Z$

$b_1 = \mu_1 t_1$

μ_1 = mass absorption coefficient for shield

t_1 = thickness of shield

μ_s = absorption coefficient of the source

Because the diameter of the fuel pins is small the self-absorption distance is small compared to the shield thickness, and

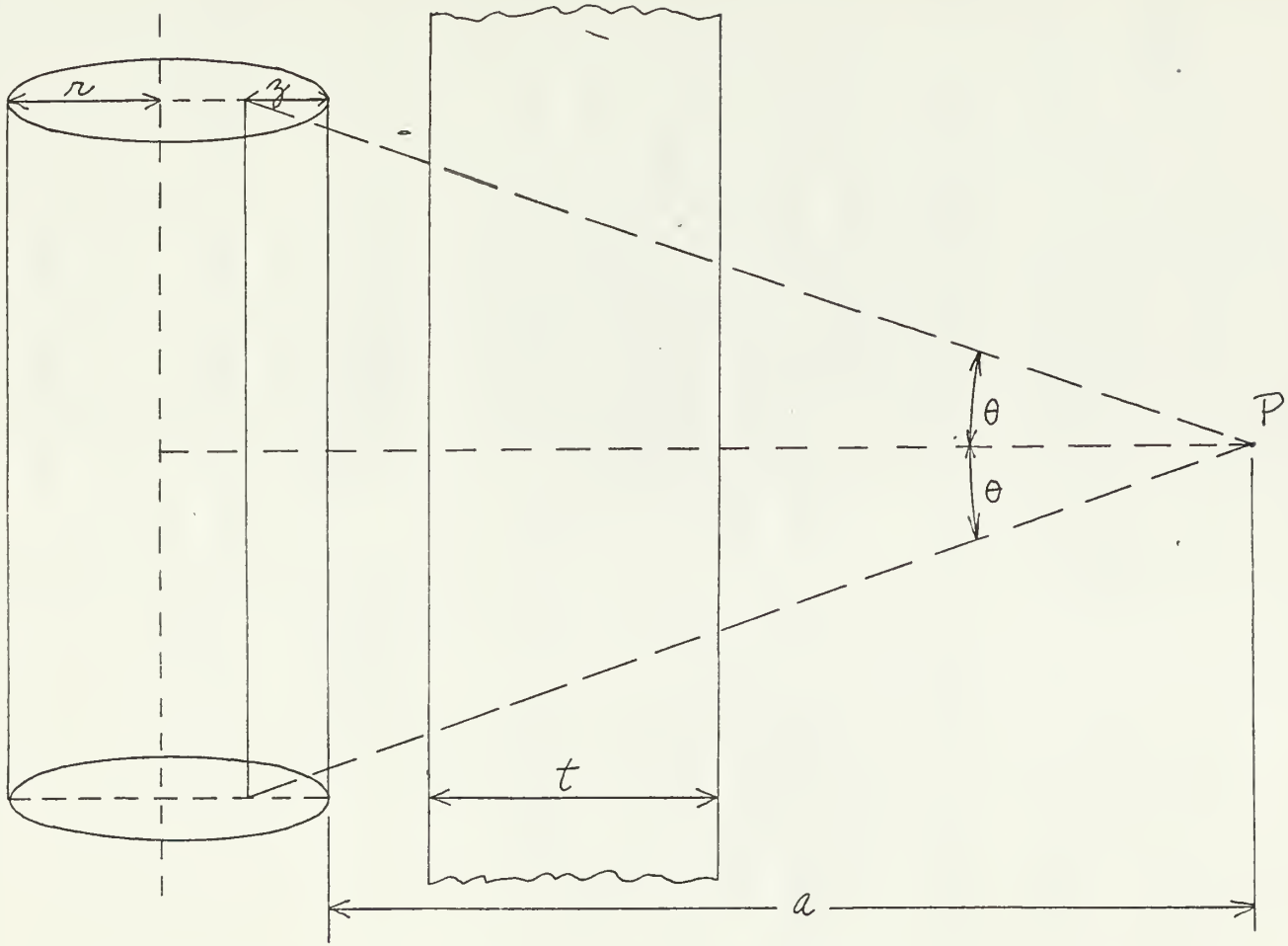

FIGURE 1 GEOMETRY for CYLINDRICAL RADIATION SOURCE

the fuel pins may be approximated as line sources of finite length.

In this case, Equation (1) simplifies to:

$$\phi = B \frac{S_L}{2\pi a} \left[F(\theta, b_1) \right] \qquad (2)$$

where:

S_L = line source strength (γ rays/cm)

In Table 4 the factors μ_1, b_1, B, $F(\theta, b_1)$, $BF(\theta, b_1)$ and $\frac{BF(\theta, b_1)}{2\pi a}$ are tabulated for a 4 foot thick water shield for each of the energy groups. Values for μ_1 and $F(\theta, b_1)$ were obtained from Reference (13). Values from B were obtained from Reference (14) for a point isotropic source in water. The value of θ used was 40°.

In Table 5 column 2, the values of the line source strengths for each group are tabulated, having been calculated from the relation:

$$S_L = \frac{\text{Activity}}{\text{Pin Length}} = \frac{\text{Activity}}{40(2.54)} \qquad (3)$$

The gamma fluxes received at the point 4 feet from the fuel pin were then calculated from Equation (2) and the values are contained in column 3 of Table 5. The flux necessary for 1 mr/hr obtained from Reference (15) is tabulated in column 4 of Table 5 for each energy group. The dose in mr/hr for each

TABLE 4: ATTENUATION FACTORS FOR 4 FT. WATER SHIELD

ENERGY GROUP No.	$E\gamma$	μ_1	b_1	$F(\theta,b_1)$	B	$BF(\theta,b_1)$	$\frac{BF(\theta,b_1)}{2\pi a}$
1	.125	.1580	19.2	1.2E-9	1000.0	1.2E-6	1.57E-9
2	.30	.1180	14.25	14.25	320.0	5.76E-5	7.51E-8
3	.50	.0975	11.90	2.0E-6	110.0	2.20E-4	2.86E-7
4	.70	.0840	10.25	1.1E-5	49.0	5.39E-4	7.03E-7
5	.90	.0750	9.14	3.4E-5	26.0	8.85E-4	1.15E-6
6	1.10	.0674	8.21	8.6E-5	19.0	1.63E-3	2.12E-6
7	1.30	.0618	7.54	1.70E-4	17.0	2.89E-3	3.77E-6
8	1.50	.0578	7.05	2.80E-4	12.0	3.36E-3	4.37E-6
9	1.70	.0540	6.58	4.70E-4	9.0	4.23E-3	5.51E-6
10	1.90	.0508	6.20	7.0E-4	7.4	5.18E-3	6.75E-6
11	2.10	.0480	5.85	1.0E-3	6.5	6.50E-3	8.47E-6
12	2.30	.0458	5.57	1.4E-3	6.3	8.82E-3	1.15E-5

TABLE 5: CONVERSION OF GAMMA SPECTRA TO DOSE RATE

ENERGY GROUP NO.	S_L	$\phi\gamma$	$\phi\gamma$ = 1 mr/hr	DOSE RATE (mr/hr)
1	1.472E10	23.12	5.0E3	.005
2	2.76E7	2.08	1.8E3	.001
3	2.42E9	692.0	1.0E3	.692
4	1.792E10	12600.0	7.2E2	17.5
5	6.72E7	77.4	5.0E2	.155
6	1.788E8	382.0	4.0E2	.995
7	7.24E6	27.4	3.8E2	.072
8	2.94E8	1286.0	3.6E2	3.572
9	2.10E7	116.0	3.4E2	.341
10	1.02E7	68.8	3.2E2	.215
11	6.96E8	5900.0	3.0E2	19.660
12	2.06E7	236.0	2.7E2	.437
			TOTAL DOSE RATE =	43.605 mr/hr

group is tabulated in column 5 of Table 5. The total dose is the sum of the doses from each energy group.

Calculations were also performed for a 2 foot thick water shield and in 1 meter of air. The dose rates for fuel pin A4 were calculated from those of A28 by multiplying by the ratio of the burnup received. The results of these calculations are summarized in Table 6.

In order to check the validity of the calculations a comparison was made between values of dose rate calculated for 2 MITR fuel elements and observed values for these elements. The results are tabulated in Table 7. It was felt that the discrepancy was due in part to the approximation of the MITR element as a line source. Because of the relatively large size of these elements appreciable attenuation would occur in the element. To check this assumption the calculation was performed again for MITR element 2M15 using the finite cylinder expression, Equation (1). The radius was assumed equal to one half the thickness of the element, 1.5 inches. The calculated dose rate using Equation (1) was 37.2 mr/hr compared to 43.6 mr/hr for the line source expression. Another reason for the discrepancy between the calculated and observed doses for the MITR elements is the intermittent operation of the M.I.T. Reactor, since Prawitz (7) assumed continuous operation in the development of the relations used in these calculations.

TABLE 6: CALCULATED DOSE RATES FROM FUEL PINS (MR/HR)

FUEL PIN	1 METER AIR	2 Ft. H_2O	4 Ft. H_2O
A4	12,420	3793	38.6
A28	14,011	4280	43.6

TABLE 7: COMPARISON OF CALCULATED AND MEASURED DOSE RATES FOR MITR FUEL ELEMENTS, 4 FT. H_2O SHIELD

FUEL ELEMENT	TOTAL[a] ENERGY MWH	IRRADIATION[a] PERIOD	COOLING[a] PERIOD	CALCULATED DOSE RATE (mr/hr)	MEASURED DOSE RATE (mr/hr)
2M15	1296	2 yrs.	850 days	104	26
2M49	1026	1.5 yrs.	130 days	1455	450

(a) Reference (21).

Because of the reasonably good agreement between the calculated and observed dose rate values for the MITR fuel it was felt the parallel calculations for the Dresden fuel would also yield adequate and even more important, conservative estimates of their expected dose rates. In particular, it should be noted that the expected dose rates from the Dresden fuel pins are well below dose rates now experienced from MITR fuel elements under similar shielding conditions. This is an important conclusion because it implies that safe handling of the Dresden fuel can be accomplished using available MITR facilities and fuel handling procedures. It is therefore recommended that licensing approval be requested from the AEC on this basis.

In view of this conclusion, it was felt that no further work was required on this aspect of the problem, and attention could therefore be focussed on the actual spectrometry, as is to be discussed in the following chapter.

III: EXPERIMENTAL INVESTIGATIONS

3.1 Introduction

In order to use gamma ray spectra for analysis of spent fuel, certain fission product gamma rays must be resolvable both in intensity and in energy so that the measured count rate is a statistically significant measure of the activity due solely to the particular fission product. In order to use the method of analysis developed by Sovka (5) it is desirable to be able to resolve the gamma rays of Cs-137, Cs-134, Zr-95 and Pr-144. These fission products have high fission yields and relatively high energy gamma rays; pertinent data concerning these fission products is presented in Table 8.

The experiments described below were performed on MITR spent fuel in order to:

1) Obtain data from which to estimate whether the gamma rays for the above fission products will be resolvable for cooling times on the order of 2 1/2 years for the power reactor spent fuel pins described in Chapter 2.
2) Determine the improvements in resolution of the spent fuel gamma spectra resulting from the improved resolution of present Ge(Li) detectors, and use of the 4096 channel analyzer, and thus ascertain if additional fission product gamma rays were suitable for analysis.

TABLE 8: PROPERTIES OF FISSION PRODUCTS USEFUL FOR GAMMA-RAY SPECTROSCOPIC STUDIES OF IRRADIATED FUEL

FISSION PRODUCT	GAMMA RAY ENERGY MeV	HALF LIFE	FISSION YIELD, %				
			THERMAL FISSION		FAST FISSION		
			U-235	Pu-239	U-235	Pu-239	U-238
Zr-95 Nb-95	0.724,0.758 0.766	65 days 35 days	6.27	5.06	6.8	5.3	5.7
Ru-106 Rh-106	2.66,2.40,2.10,1.55 1.05,0.624,0.607,0.513	1.0 yr 30 sec.	0.38	4.04	0.4	6.3	2.7
Cs-134 Cs-133	0.605,0.796 None	2.3 yr. Stable	6.59	Cs-133(n,γ) 5.59	Cs-134	-	-
Cs-137	0.662	30 yrs.	6.00	5.40	6.2	5.8	6.2
Ce-144 Pr-144	0.079,0.133 0.697, 1.488, 2.186	285 days 17 min	5.62	4.09	4.8	3.7	4.9

Reference (5)

3) Check operation of the scanning system and make alterations as necessary to enable the equipment to scan and hold spent power reactor fuel pins.

3.2 Experimental Equipment and Procedures

3.2.1) MITR Fuel Elements

MITR fuel elements are of the standard MTR plate type construction. A detailed description is contained in Reference (5). The important feature for present purposes is that the elements contain approximately 160 grams before irradiation of highly enriched uranium (93% U-235). Gamma ray spectra were recorded for two elements, 7M9 and 5M22. Some of the pertinent information for the elements are listed in Table 9.

3.2.2) Scanning Equipment and Procedures

Scanning equipment developed by Sovka (5) for scanning spent MITR fuel was used to obtain spectra from irradiated MITR fuel. A schematic diagram, shown in Figure 2, illustrates the major features of the system which allows the scanning of spent fuel to be accomplished in the MITR spent fuel storage pit. The detector dewar is positioned on the upper movable carriage and is surrounded by lead shielding to reduce background. The upper movable carriage is positioned on the lower movable carriage, which permits scanning in two perpendicular directions in the horizontal plane. Extending from the upper carriage through the water to the fuel element

TABLE 9: IRRADIATED FUEL ELEMENT DATA

FUEL ELEMENT	DATE CHARGED	DATE DISCHARGED	TOTAL ENERGY, MWH	ORIG. U-235 Wt., gms	FINAL U-235 Wt., gms	COOLING TIME
5M27	10/12/67	8/9/68	948.1	161.19	112.98	272 Days
7M9	3/14/68	2/22/69	1079.7	158.83	101.26	79 Days

Reference (21).

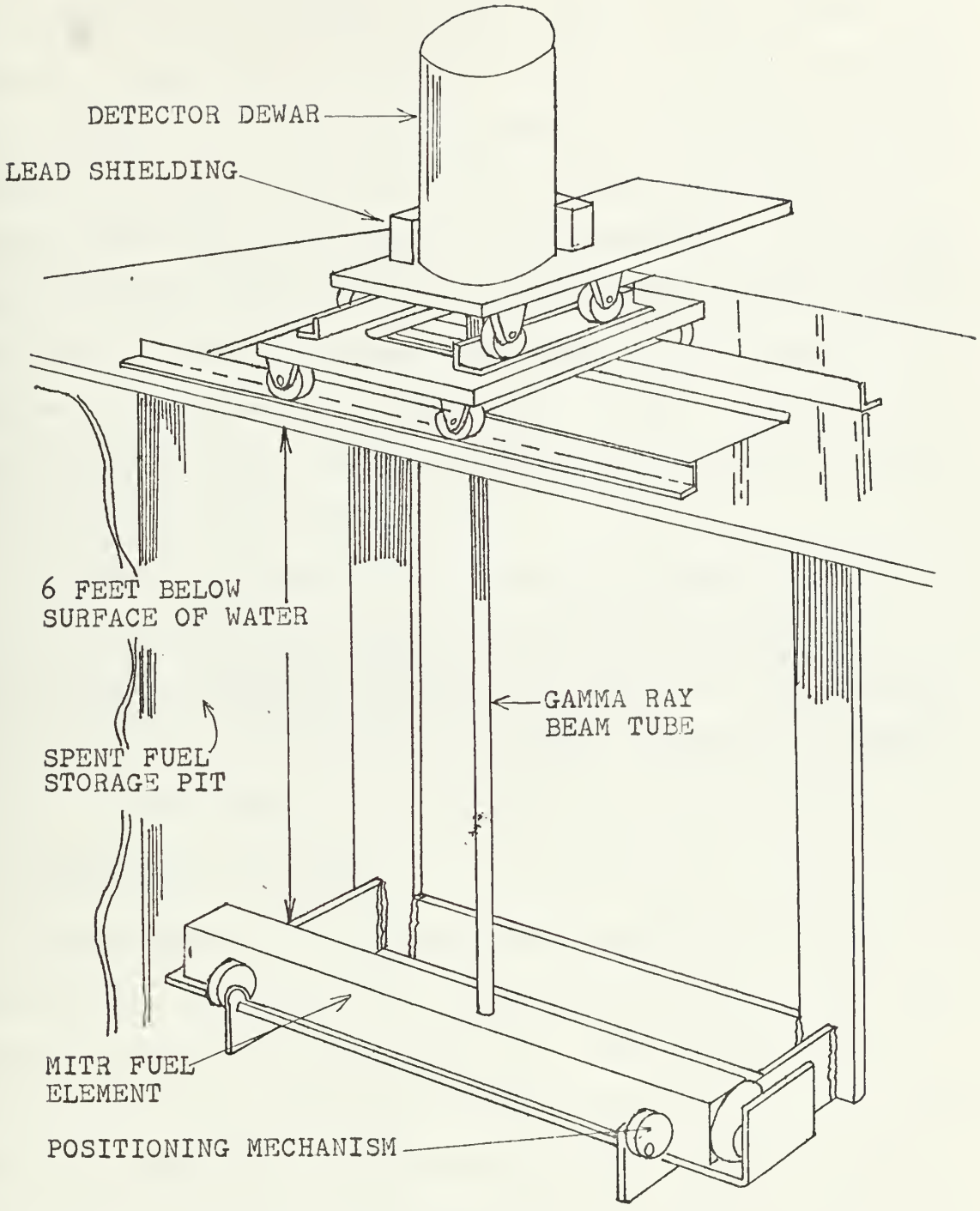

FIGURE 2 SCHEMATIC DIAGRAM of APPARATUS for GAMMA RAY SCANNING of SPENT FUEL ELEMENTS

is a 1/2 in. I.D. air filled tube which acts as a collimator and allows a well-collimated beam of gamma rays to reach the detector. Additional collimators made of lead with diameters from 1/2 in. to 1/8 in. may be placed in the beam of the top carriage to reduce the count rates to acceptable levels and provide additional shielding against background. In the present experiment, lead sheets were also placed in the beam to absorb many of the low energy gamma rays in the beam while allowing the higher energy gamma rays to pass through relatively unattenuated. Count times of 80 to 160 minutes were used depending on the count rate.

A schematic diagram of the electronic components used together with important settings are shown in Figure 3. A list of these components is shown in Table 10. The settings shown in Figure 3 resulted in an energy decrement of approximately .66 keV/channel.

3.3 Experimental Results

Identification of the photopeaks noted in the spectra obtained from the spent fuel elements was made utilizing the energy calibration of Reference (5). The improved resolution of the Ge(li) detectors now manufactured at M.I.T. allowed better resolution of the spectra, while their increased size and hence greater efficiency made shielding against background more difficult. Orginally, a detector with an active volume of 30 cm^3 was used, but even with 4 inches of lead

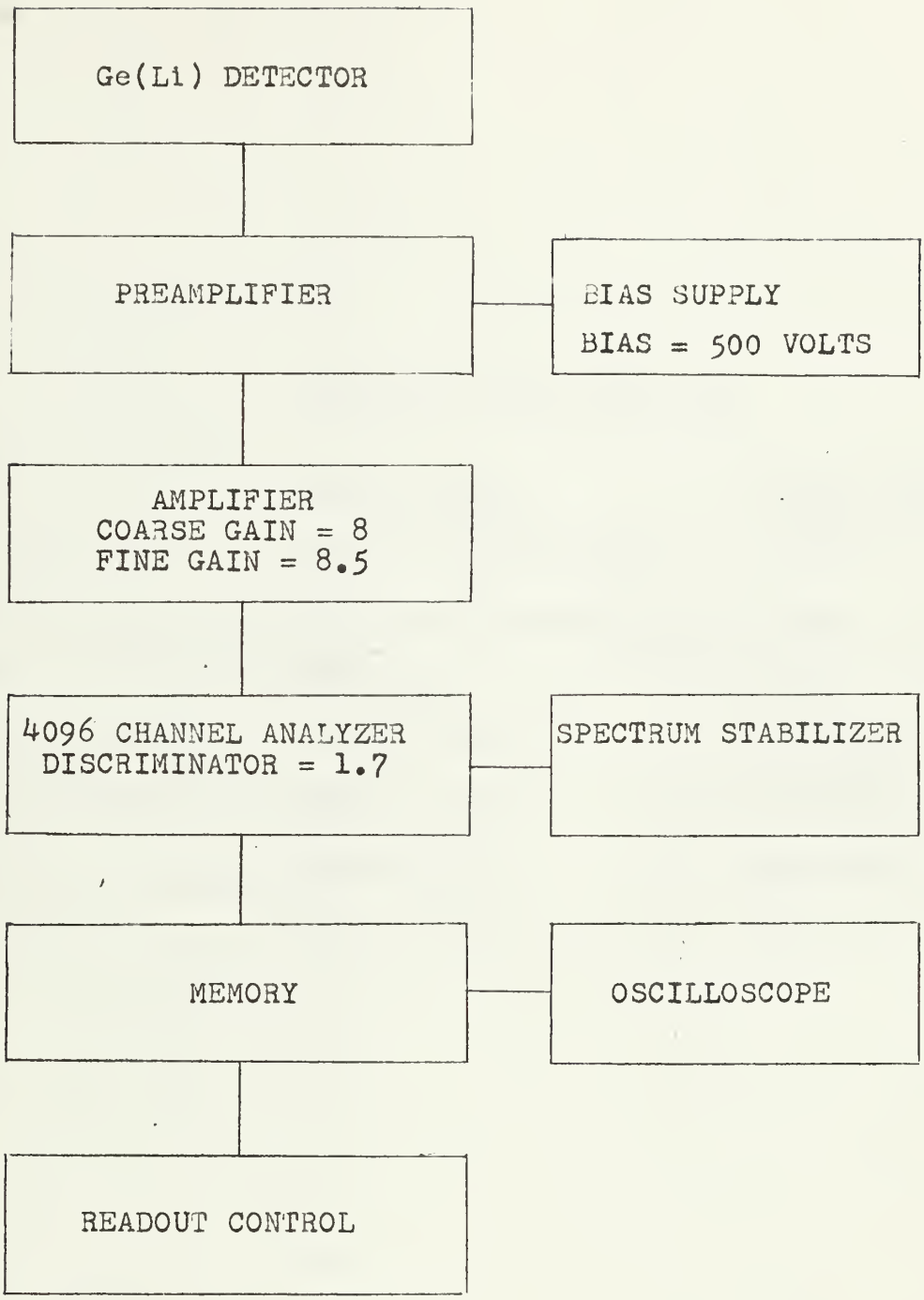

FIGURE 3 SCHEMATIC DIAGRAM of ELECTRONICS USED for OBTAINING GAMMA RAY SPECTRA FROM SPENT MITR FUEL

TABLE 10: ELECTRONIC EQUIPMENT DATA

EQUIPMENT	MANUFACTURER	MODEL NO.
Preamplifier	Canberra Instruments	1408C
Bias Supply	Canberra Instruments	3001
Amplifier	Sturrup	1417
4096 Channel Analyzer	Nuclear Data	ND-161F
Spectrum Stabilizer	Nuclear Data	ND-502
Memory Unit	Nuclear Data	ND-160M
Readout Control	Nuclear Data	ND-160R

shielding on the bottom of the detector and 2 inches of lead shielding on the side of the detector, dead times of 30-35% were noted even with the collimator opening also shielded by 4 inches of lead. Additional shielding could not be added as the amount of shielding material was limited by the mechanical strength of the scanning equipment. This larger detector was replaced with a smaller detector having an 8 cm^3 volume. The resulting dead times were found to be on the order of 30% with the collimator tube open and an additional 1/8 in. aperture collimator inserted immediately below the detector when element 7M9 was placed in the holder.

3.3.1) Fission Product Gamma Ray Spectra

3.3.1a) Fuel Element 7M9

The spectrum obtained from MITR fuel element 7M9 after a cooling period of 79 days is shown in Figure 4. The spectrum was obtained with a 1/8 in. diameter aperture collimator for a counting time of 80 minutes. In addition three 1/4 in. lead sheets were placed in series in the beam to preferentially reduce low energy gamma rays in the beam. The spectrum shows resolved peaks for gamma rays from the decay of Nb-95, Pr-144, Cs-134 and Cs-137. Except for the peaks due to Pr-144, all resolved peaks lie between 500 and 800 keV, and an enlargement of this portion of the spectrum is shown in Figure 5.

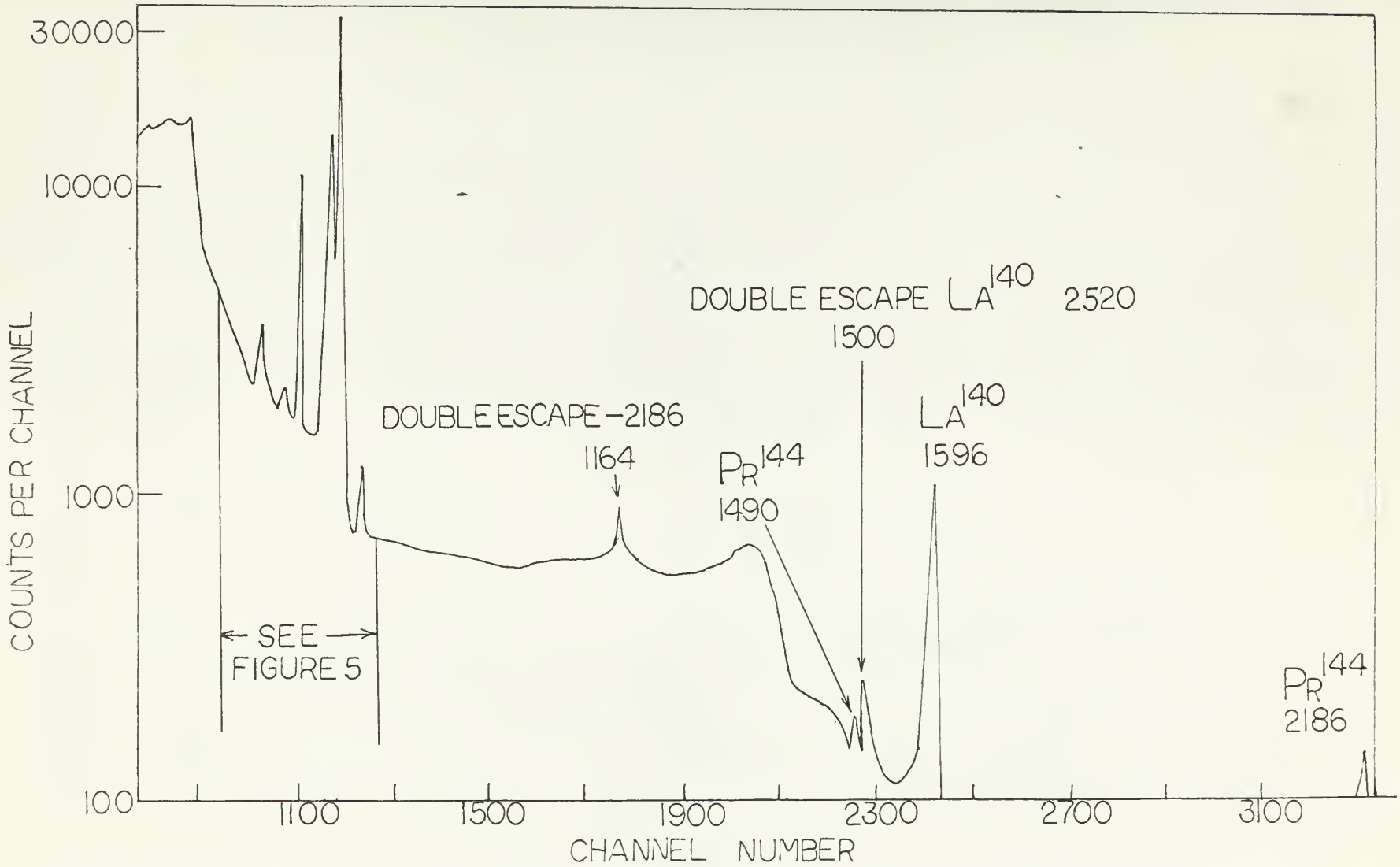

FIGURE 4 GAMMA RAY SPECTRUM (500-2200 KEV) OF MITR FUEL ELEMENT 7M9 AFTER 79 DAYS COOLING

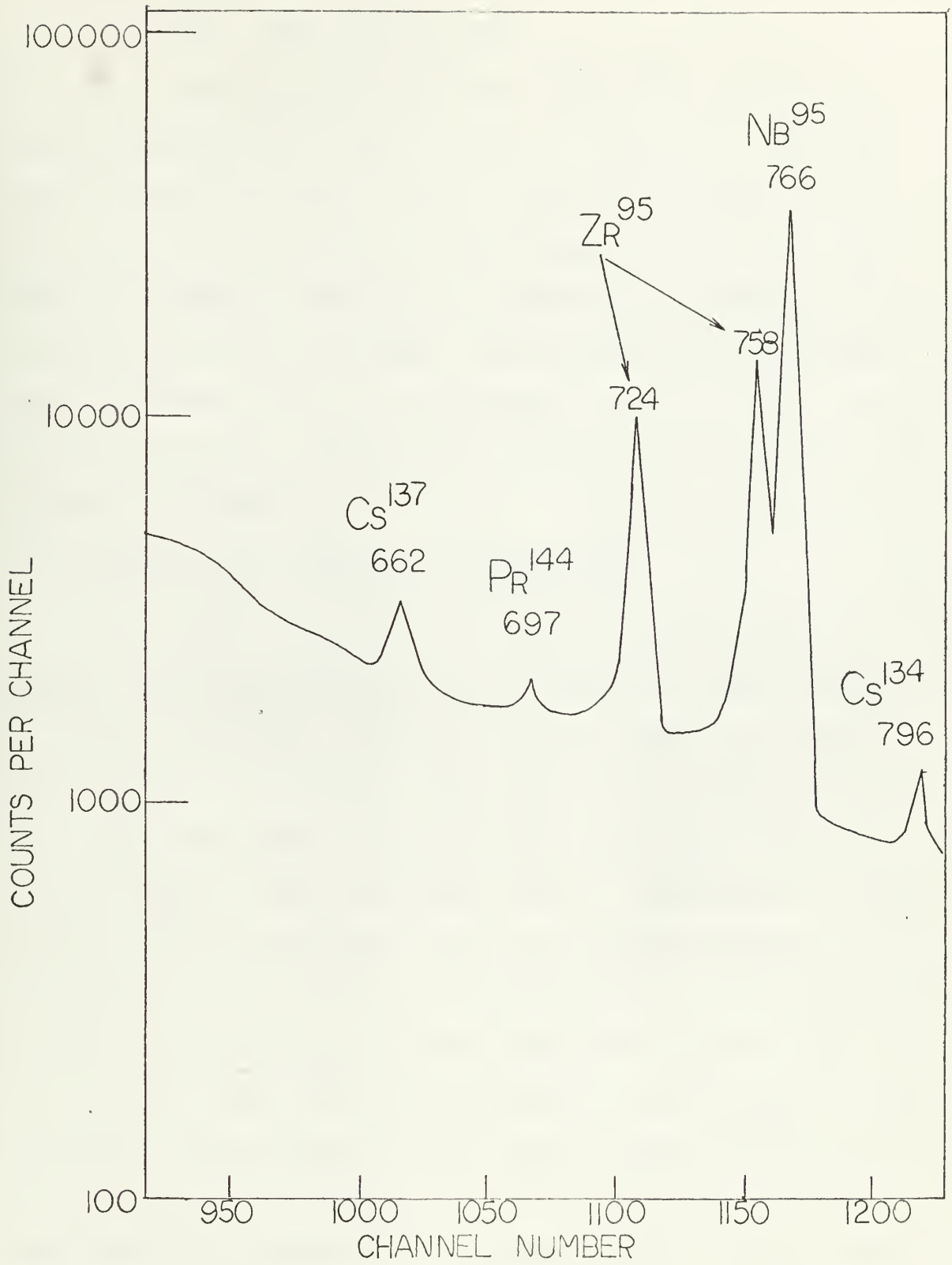

FIGURE 5 GAMMA RAY SPECTRUM (600–800KEV) OF MITR FUEL ELEMENT 7M9 AFTER 79 DAYS COOLING

3.3.1b) Fuel Element 5M27

The spectrum from MITR fuel element 5M27 after a cooling period of 272 days is shown in Figure 6. The spectrum was obtained using a 1/8 in. diameter aperture collimator for a counting time of 160 minutes. One 1/4 in. lead sheet was placed in the beam to preferentially reduce the low energy gamma rays in the beam. The spectrum shows resolved peaks for gamma rays due to the decay of Nb-95, Zr-95, Pr-144, Cs-134 and Cs-137. The spectrum between 600 keV and 800 keV is shown in Figure 7.

3.4 Calculations of Gamma Ray Intensities

Intensities of gamma rays resolved in the spectra of Figures 4 and 6 were calculated by finding the area, expressed in terms of counts, under each peak. These calculations were performed using GAMANL, Reference (16), a computer code which identifies peaks in spectra and determines their centers and areas. GAMANL utilizes Fourier transforms to smooth the data, which eliminates much of the random fluctuations in the spectra without affecting the resolution. The background is then calculated and subtracted from the smoothed data and the resulting peaks are then analyzed for peak centers, areas and FWHM's, among other things. The areas are calculated in two ways, in the first method all the counts are added between successive zeroes in the smoothed data with background

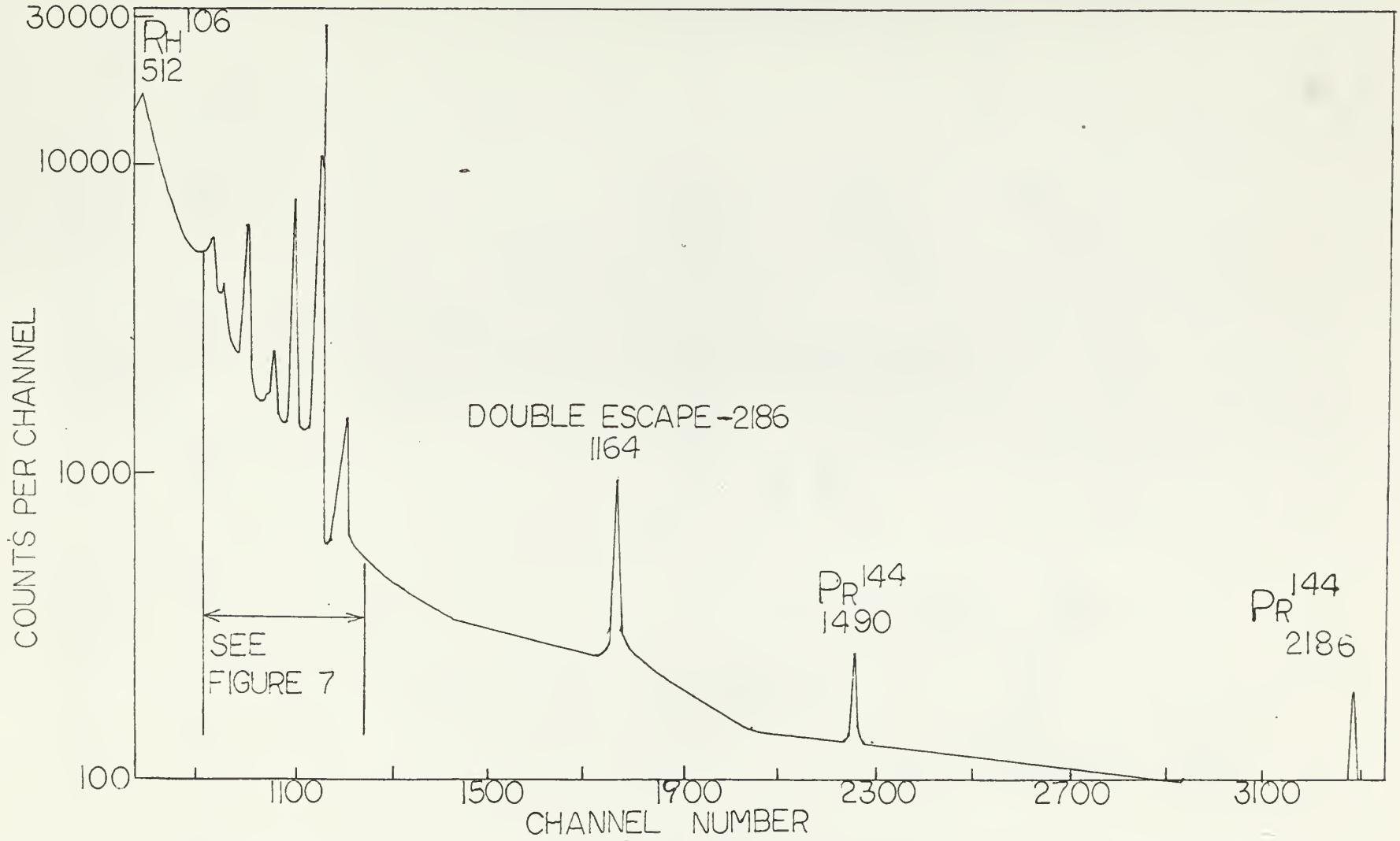

FIGURE 6 GAMMA RAY SPECTRUM(500-2200'KEV) OF MITR FUEL ELEMENT 5M27 AFTER 9 MONTHS COOLING

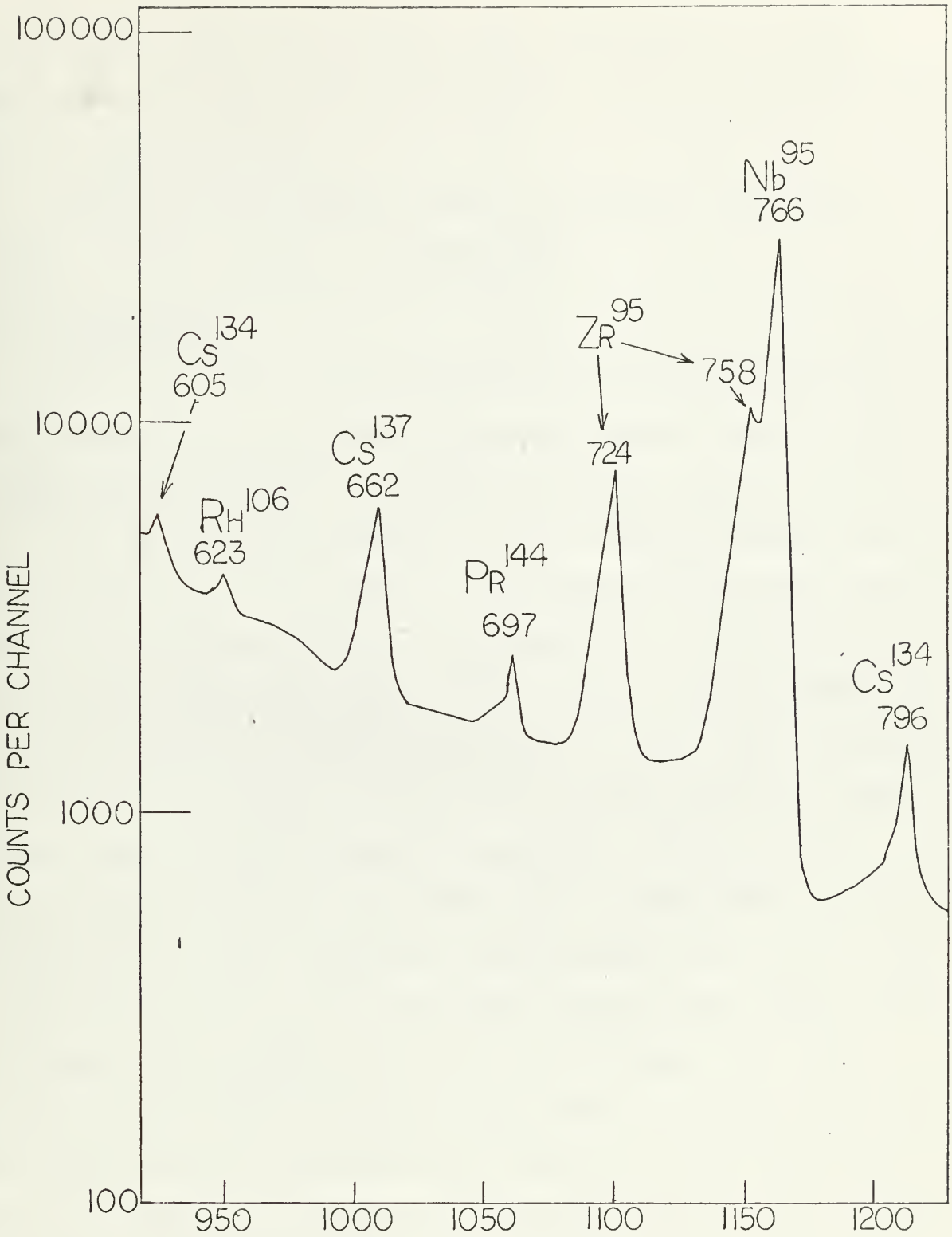

FIGURE 7 GAMMA RAY SPECTRUM (600-800 KEV) OF MITR FUEL ELEMENT 5M27 AFTER 9 MONTHS COOLING

subtracted. The second method uses a Gaussian fit to the peaks and calculates the area using the height of the peak and its standard deviation.

Results for the spectra shown in Figures 4 and 6 are shown in Tables 11 and 12 respectively.

3.5 Conclusions

Based upon the results obtained in the work reported in this chapter, the following conclusions can be drawn pertinent to the projected future experiments on power reactor fuel:

1) The improved resolution of the Ge(Li) detectors now manufactured at M.I.T. and the 4096 channel and analyzer are evident in partial resolution of the Zr-95 Nb-95 doublet at 758 and 766 keV. This consideration is not too important for cooling times on the order of one year or less since the peaks necessary for analyses are all strong peaks and tend to mask weaker peaks in their Compton background. After several years cooling, however, the peaks due to gamma rays from Pr-144 and Cs-134 will have decreased considerably due to their relatively short half lives of 285 days and one year respectively. The peaks due to Nb-95 and Zr-95 will decrease even more rapidly due to their effective half life of 65 days, and will be a relatively small peak after a cooling time on the order of 1 1/2 years. The activity of fission products with longer half lives, although having lower fission yields, will begin to approach the activity of Zr-95, Nb-95, Pr-144 and Cs-134,

TABLE 11: CALCULATED PEAK AREAS FOR MITR FUEL ELEMENT 7M9 SPECTRUM

FISSION PRODUCT	GAMMA RAY ENERGY (keV)	PEAK AREA METHOD (A) (Counts)	PEAK AREA METHODS (B) (Counts)	ERROR METHOD (B) %
Pr-144	2186	1025.2	854.5	19.78
	1488	425.9	464.8	16.89
	1164.4[a]	2275.3	2550.2	7.23
Cs-134	796	1682.1	1799.1	8.35
	605	Not Resolved		
Nb-95	766	239,556.0	218,766.2	1.15
Zr-95	758	92,502.5	85,345.4	1.23
	724	62,367.5	57,377.1	1.39
Cs-137	662	7,168.4	7,041.0	4.13
Ru-106	511	4,588.6	3,442.0	15.83

(a) Double Escape Peak

TABLE 12: CALCULATED PEAK AREAS FOR MITR FUEL ELEMENT 5M27 SPECTRUM

FISSION PRODUCT	GAMMA RAY ENERGY (keV)	PEAK AREA METHOD (A) (Counts	PEAK AREA METHOD (B) (Counts)	ERROR METHOD (B) %
Pr-144	2186	2441.2	1914.0	15.97
	1488	950.2	1034.5	10.40
	1164[a]	5658.8	5884.4	4.40
Cs-134	796	9060.9	8003.9	2.92
	605	7787.4	8387.7	5.41
Nb-95	766	240387.9	228593.6	1.52
Zr-95	758	84910.4	80577.3	1.60
	724	52532.8	48988.5	1.66
Cs-137	662	32145.6	30966.5	2.06
Ru-106	622	2326.8	3225.9	2.88
	511	6918.1	7435.1	10.01

(a) Double Escape Peak

and the resolution of the system becomes considerably more important.

2) In the future, the discriminator setting should be set higher (e.g. at 400 keV) than it was for these runs (52 keV) to help reduce counting system dead times. Because it was desirable to minimize the need for movement of the detector, the first fuel element was placed in the holder prior to placing the dewar containing the detector on the top carriage. This made it impossible to make background runs prior to obtaining the original spectra, and peaks noted in the channels below 900 were at one time thought to be significant. Subsequent background runs proved this original assumption to be incorrect. With MITR fuel element 7M9 in the holder but with the collimator tube removed, dead times on the order of 15% were noted for a discriminator setting of 1.7, corresponding to a cut off energy of 52 keV. For a discriminator setting of 4.2, corresponding to a cut off energy of 350 keV, the dead times were reduced to 3%.

3) With the reduced background that will result from the less active spent power fuel pins, it would be possible to use a larger detector, which with its improved intrinsic efficiency will increase the number of counts measured, especially for the higher energy gamma rays. The use of a larger detector should also help reduce the Compton edges somewhat by enabling more gamma rays to deposit their full energy.

4) Calibration of the system for the double escape peak of the 2186 kev Pr-144 gamma ray should also improve the determination of the Pr-144 content of the spent fuel. However, it should be noted the use of a larger detector may decrease the probability of both gamma rays escaping from the detector after a Pair production event and thus decrease the size of the double escape peak.

In short, it is concluded that the proposed power fuel experiments using the apparatus described in this chapter are quite feasible. Sufficient signal strength is easily obtained, only 1/16th of the full collimator bore was used in the present scoping studies. Room background can be reduced to acceptable levels by proper choice of detector size, lead filter thickness and shielding. Dead time can be reduced by using a sufficiently high low level discriminator cut off. The key limitation is really in the Compton background produced in the detector.

So far, then, we have investigated and established the potential ability to handle spent fuel and carry out a spectrometer scan with available apparatus. In the next chapter a projection will be made concerning the suitability of the data obtainable with this apparatus for the fuel in question.

IV: CALCULATION OF GAMMA RAY INTENSITIES EXPECTED IN SPENT FUEL.

4.1 Introduction

In order to determine the feasibility of studying the spent fuel made available by the AEC it was necessary to determine the expected intensities of those gamma rays useful for analysis. It was also necessary to determine whether they would be resolvable for a cooling time of approximately 2.75 years. In addition to calculating the gamma rays emitted by fission products of U-235 fissions, an estimate was made of the expected gamma rays from fission products of Pu-239.

4.2 Calculation of Expected U-235 Fission Product Gamma Ray Intensities.

Calculation of the expected intensities for gamma rays from U-235 fission products was performed by utilizing the calculated peak area (expressed as counts) obtained in Chapter 3. The data obtained for MITR fuel element 5M27 was extrapolated, using the familiar experimental decay relationships, to a 2.75 year cooling period, which is the expected cooling period for the Dresden fuel pins. The calculated intensities are shown in Table 13.

In order to check the validity of extrapolating the intensity of a gamma ray, as measured by the area under its peak, on the gamma ray spectra, a parallel calculation was performed extrapolating the measured intensity for the gamma rays of 7M9 (cooling time = 2.75 months) to a cooling period of 9

TABLE 13: INTENSITY OF U-235 FISSION PRODUCT GAMMA RAYS AFTER 2.75 YEARS COOLING

FISSION PRODUCT	ENERGY (keV)	PEAK AREA
Pr-144	2186	322.8
	1490	174.5
	1164	992.4
	696	1,102.2
Cs-134	796	4,129.3
	605	4,327.4
Nb-95	766	93.3
Zr-95	758	32.9
	724	19.9
Cs-137	662	29,565.7
Rh-106	623	544.0
	511	1280.0

months. Corrections were made for the differences in counting time, thickness of lead sheets in the beam, and burnup. These calculated intensities are compared with the measured intensity (peak area) for the gamma rays of 5M27 (cooling time = 9 months) in Table 14.

The calculated intensities compare well with the measured intensities with the exception of Cs-137. No reason for the discrepancy could be discovered since both 5M27 and 7M9 had similar irradiation histories and were used in similar positions in the MITR core.

In order to determine if the values of the calculated intensities listed in Table 13 were sufficiently strong to be resolved, their relative intensities were analyzed. This method of analysis was chosen because with spent fuel gamma spectra the difficulty is not one of obtaining sufficient count rate but rather of resolving lower energy gamma rays in the presence of Compton background from higher energy gamma rays. Since the height of the Compton edge and continuum are proportioned to the size of the peak it was felt that the ratio of a gamma ray's peak area to that of the sum of the peak areas for higher energy gamma rays would yield sufficient information to predict which gamma rays would be resolvable. The results of this analysis are shown in Table 15. For comparison the results of the GAMANL (16) calculation for fuel element 5M27 were analyzed in this manner and the results are listed in Column 3 of Table 15. From this it was concluded

TABLE 14: CALCULATED AND MEASURED INTENSITY OF U-235 FISSION PRODUCT GAMMA RAYS AFTER 9 MONTHS COOLING.

ENERGY (keV)	CALCULATED PEAK AREA	MEASURED PEAK AREA
2186	1,836.9	1914.0
1490	1,105.3	1034.5
1164	6,904.4	5884.4
796	8,880.6	8003.9
766	178,525.2	228,593.6
758	69,640.8	80,577.3
724	46,819.0	48,988.5
696	6,980.4	6,535.3
662	53,573.6	30,966.5

TABLE 15: CALCULATED VALUES OF RELATIVE INTENSITY FOR U-235 FISSION PRODUCTS AFTER 2.75 YEARS COOLING.

ENERGY (keV)	CALCULATED RELATIVE INTENSITY	MEASURED RELATIVE INTENSITY (9 mos. cooling)
2186	-	-
1490	.54	.54
1164	1.99	1.99
796	8.30	2.72
766	.02	20.70
758	.0069	.337
724	.0042	.153
696	.227	.017
662	5.03	.082
623	.015	.008
605	.106	.020
511	.030	.017

that all gamma rays were capable of being resolved by use of GAMANL (16). However, it is felt that the low absolute intensity of the peaks at 766, 758 and 724 keV would not lead to reproducible results in the calculation of the areas under the peaks.

4.3 Calculation of Expected Pu-239 Fission Product

4.3.1 Gamma Ray Intensities

In order to obtain an estimate of the number of Pu-239 fissions which have occurred in the fuel pins an energy balance was performed using power history data supplied by General Electric for the fuel in question. It was assumed that the energy was released either by thermal fission of U-235 or Pu-239, i.e.:

$$E(\text{TOTAL}) = E(\text{Pu-239}) + E(\text{U-235})$$

Assuming 200 Mev are released per fission:

$$15.3\text{g U-235} \times \frac{6.023 \times 10^{23} \text{ atoms}}{\text{gm - mole}} \times \frac{\text{gm mole}}{235 \text{ gm U-235}} \times \frac{1 \text{ fission}}{1 \text{ atom}}$$

$$\times \frac{200 \text{ Mev}}{\text{fission}} \times \frac{577b}{683b} \frac{\text{fission U-235}}{\text{fission + capture U-235}} \times \frac{\text{watt - sec}}{6.25 \times 10^{12} \text{ Mev}}$$

$$\times \frac{\text{day}}{8.64 \times 10^{4} \text{ sec}} \times \frac{\text{MW}}{10^{6} \text{ watt}} = \frac{15.3(6.023)(577)(200)}{235(683)(8.64)(6.25)} = 12.2 \text{ MWD}$$

Thus:

$$E(\text{Pu-239}) = E(\text{TOTAL}) - E(\text{U-235})$$

$$= 17.08 - 12.20$$

$$= 5.08 \text{ MWD}$$

$$\text{The ratio of } \frac{\text{fissions in Pu-239}}{\text{fissions in U-235}} = \frac{5.08}{12.2} = .416$$

Since approximately 42% of the fissions are in Pu-239 there is a possibility of resolving Pu-239 fission product gamma rays in the spectrum. The value of 42% is in good agreement with the values predicted using cross section data as reported in Appendix A.

From Table 8 it is seen that the fission yield of Rh-106 - Ru-106 is increased from 0.4% for thermal fission of U-235 to 4.4% for thermal fission of Pu-239. Table 13 shows the predicted peak area for the 623 keV peak of Rh-106 is 544.0 counts for fission of U-235. The peak area for fission of Pu-239 would be

$$\frac{4.4\%}{.4\%} \times 544.0 = 5440.0 \text{ counts}$$

The superimposed peak area for the fuel pins for the 623 keV gamma ray would then be

$$.416(5440.0) + .584(544.0) = 2574.0 \text{ counts}$$

This is an appreciable percentage of the peak area for Cs-137 and thus should be resolvable.

4.4 Conclusions

A sufficient number of gamma ray peaks should be resolvable and statistically significant in the spectra to enable

nondestructive testing to be performed. Only one gamma ray need be resolved for each fission product. Even though the gamma rays of Nb-95 and Zr-95 will not be resolvable, the gamma rays of Rh-106 at 623 keV should be resolvable and thus the method of analysis utilizing three fission product activity ratios recommended by Sovka (5) should be possible.

V: CONCLUSIONS

The major conclusions reached as a result of the work reported in this thesis are as follows:

1) The calculated dose rate for the Dresden power reactor fuel pins is well below that presently experienced from spent MITR fuel elements, and thus presents no special handling problems.

2) Sufficient spectral information should be obtained from the spent Dresden fuel pins to enable non-destructive analysis to be performed. In fact, the increased energy resolution offered by present day Ge(Li) detectors and the 4096 channel analyzer coupled with the improved analytic capability of the GAMANL code (16) allow improved determination of the actual gamma ray intensities from spent fuel spectra as compared to previous studies on MITR fuel by Sovka (5).

3) Although Nb-95 and Zr-95 gamma rays will probably not be resolved in the subject Dresden fuel, the expected relative increase in Ru-106 and Rh-106 will permit substitution of this latter pair in the analytic procedure.

4) A method of Compton suppression would be extremely valuable because of the intense Compton background present between 600 and 800 keV, the energy range for all but one of the gamma rays required for the method of analysis presented by Sovka (5). In addition, Compton

4) cont.

suppression might enable weaker intensity gamma rays potentially useful for analysis to be resolved. The methods utilizing anti-coincidence would be extremely difficult to apply because of volume and weight limitations imposed by the scanning equipment. A duode type Ge(Li) detector (17,18), however, would be ideally suited to scanning of spent fuel since it requires no additional space or volume than that needed for a standard Ge(Li) detector.

As a result of this work, it can be said that all necessary groundwork for spent power reactor fuel pin analysis has been completed to the point where shipping and receipt of the fuel, and initiation of scanning experiments is now warranted.

APPENDIX A

A parallel calculation of the ratio of the number of fission in Pu-239 to the number of fissions in U-235 was carried out using one group theory utilizing effective neutron cross sections to account for the fission caused by epithermal neutrons and to account for all neutron captures in U-238.

In order to obtain an expression for the number of Pu-239 fissions we must first obtain an expression for the number of U-238 atoms converted to Pu-239 atoms as a function of time. For the one group model we can write:

$$\frac{dN^{28}}{dt} = -\overline{\sigma}_{n,\gamma}{}^{28}\phi N^{28} \qquad (1)$$

where:

N^{28} = no. of U-238 atoms at time t

$\overline{\sigma}_{n,\gamma}{}^{28}$ = effective microscopic capture cross section for U-238

ϕ = one group neutron flux

Solving Eq. (1)

$$N^{28} = N_o^{28}\left[\exp(-\overline{\sigma}_{n,\gamma}{}^{28}\phi t)\right] \qquad (2)$$

where:

N_o^{28} = initial no. of U-238 atoms

For Pu-239 atoms we can write

$$\frac{dN^{49}}{dt} = \overline{\sigma}_{n,\gamma}{}^{28}\phi N^{28} - \overline{\sigma}_a^{49}\phi N^{49} \qquad (3)$$

where:

N^{49} = no. of Pu-239 atoms at time t

σ_a^{49} = effective microscopic cross section for Pu-239

Substituting Eq. (2) for N^{28} in Eq. (3)

$$\frac{dN^{49}}{dt} = \sigma_{n,\gamma}^{28} \phi N_o^{28} \left[\exp(-\sigma_{n,\gamma}^{28} \phi t) \right] - \sigma_a^{49} N^{49} \qquad (4)$$

Solving Eq. (4) yields

$$N^{49} = N_o^{49} \left[\exp(-\sigma_a^{49} \phi t) \right] + N_o \left(\frac{\sigma_{n,\gamma}^{28}}{\sigma_{n,\gamma}^{28} + \sigma_a^{49}} \right) \left[\exp(-\sigma_{n,\gamma}^{28} \phi t) - \exp(-\sigma_a^{49} \phi t) \right]$$

Since $N_o^{49} = 0$, $\sigma_a^{49} \gg \sigma_{n,\gamma}^{28}$ and $\exp(-\sigma_{n,\gamma}^{28} t) \approx 1$ for normal fuel burnup the above expression can be approximated by

$$N^{49} = N_o^{28} \left(\frac{\sigma_{n,\gamma}^{28}}{\sigma_a^{49}} \right) \left[1 - \exp(-\sigma_a^{49} \phi t) \right] \qquad (5)$$

For the time ratio of Pu-239 fissions we can write

$$\frac{dF^{49}}{dt} = \sigma_f^{49} \phi N^{49} \qquad (6)$$

Substituting Eq. (5) for N^{49} in Eq. (6) we have

$$\frac{dF^{49}}{dt} = \sigma_f^{49} \phi N_o^{28} \left(\frac{\sigma_{n,\gamma}^{28}}{\sigma_a^{49}} \right) \left[1 - \exp(-\sigma_a^{49} \phi t) \right]$$

Integration yields

$$F^{49} = \sigma_f^{49} \phi N_o^{28} \left(\frac{\sigma_{n,\gamma}^{28}}{\sigma_a^{49}} \right) \left[t - \left(1 - \exp(\sigma_a^{49} \phi t) \right) / \sigma_a^{49} \phi \right] \qquad (7)$$

Expanding the exponential term of Eq. (7) in a Taylor Series yields

$$F^{49} = \sigma_f^{49} \phi N_o^{28} \left(\frac{\sigma_{n,\gamma}^{28}}{\sigma_a^{49}} \right) \left[t - \left(1 - 1 + \sigma_a^{49} \phi t - 1/2 (\sigma_a^{49} \phi t)^2 + \ldots \right) / \sigma_a^{49} \phi \right]$$

Ignoring all terms above the t^2 term yields

$$F^{49} = \sigma_f^{49} \phi N_o^{28} \left(\frac{\sigma_{n,\gamma}^{28}}{\sigma_a^{49}} \right) (1/2 \ \sigma_a^{49} \phi t^2) \qquad (8)$$

For the number of U-235 atoms we may write

$$\frac{dN^{25}}{dt} = - \sigma_a^{25} \phi N^{25} \qquad (9)$$

where:

N^{25} = no. of U-235 atoms at time t

σ_f^{25} = effective microscopic cross section for U-235

Solving Eq. (9)

$$N^{25} = N_o^{25} \exp(- \sigma_a^{25} \phi t) \qquad (10)$$

The rate of U-235 fissions is given by

$$\frac{dF^{25}}{dt} = \sigma_f^{25} \phi N^{25} \tag{11}$$

where: F^{25} = total no. of U-235 fissions from time t =0

Substituting Eq. (10) into Eq. (11)

$$\frac{dF^{25}}{dt} = \sigma_f^{25} \phi N_o^{25} \quad (1-\exp(-\sigma_a^{25} \phi t))/\sigma_a^{25} \tag{12}$$

The ratio of Pu-239 fissions to U-235 fissions using Eq. (8) and Eq. (12) becomes

$$\frac{F^{49}}{F^{25}} = \left(\frac{\sigma_f^{49}}{\sigma_a^{49}}\right)\left(\frac{N_o^{28}\,\sigma_{n,\gamma}^{28}}{N_o^{25}\,\sigma_f^{25}}\right)\left(\frac{(1/2)(\sigma_a^{49}\phi t^2)(\sigma_a^{25}\phi)}{1-(N^{25}/N_o^{25})}\right)$$

$$= \frac{\sigma_f^{49}}{2}\left(\frac{N_o^{28}}{N_o^{25}-N^{25}}\right)\left(\frac{\sigma_{n,\gamma}^{28}}{\sigma_f^{25}}\right)\sigma_a^{25}(\phi t)^2 \tag{13}$$

In order to eliminate flux and time dependence of Eq. (13) return to Eq. (10) and take logarithms of both sides

$$\ln(N^{25}/N_o^{25}) = \sigma_a^{25} \phi t$$

Solving for ϕt yields

$$\phi t = (1/\sigma_a^{25}) \ln(N^{25}/N_o^{25})$$

Substituting for t in Eq. (8) yields

$$\frac{F^{49}}{F^{25}} = 1/2 \left(\frac{\sigma_f^{49}}{\sigma_f^{25}}\right)\left(\frac{\sigma_a^{28}}{\sigma_a^{25}}\right)\left(\frac{N_o^{28}}{N_o^{25} - N^{25}}\right)\left[\ln\left(\frac{N_o^{25}}{N^{25}}\right)\right]^2 \qquad (14)$$

Rearranging yields

$$\frac{F^{49}}{F^{25}} = 1/2 \left(\frac{\sigma_f^{49}}{\sigma_f^{25}}\right)\left(\frac{\sigma_a^{28} N_o^{28}}{\sigma_a^{25} N_o^{25}}\right)\left(\frac{1}{1-(N^{25}/N_o^{25})}\right)\left[\ln\left(\frac{N_o^{25}}{N^{25}}\right)\right]^2$$

The ratio $\sigma_a^{28} N_o^{28} / \sigma_a^{25} N_o^{25}$ is defined as the initial conversion ratio (ICR) and is equal to the initial number of neutron absorptions in U-238 divided by the number of neutron absorptions in U-235. Thus the final working equation becomes

$$\frac{F^{49}}{F^{25}} = 1/2 \left(\frac{\sigma_f^{49}}{\sigma_f^{25}}\right) \frac{ICR}{(1-N^{25}/N_o^{25})} \left[\ln\left(\frac{N_o^{25}}{N^{25}}\right)\right]^2 \qquad (15)$$

In order to apply Eq. (15) we need to specify the effective cross sections σ_f^{49} and σ_a^{49}. To account for both thermal and epithermal events the following approximate formulation can be employed:

$$\bar{\sigma} = \sigma_{th} + \left[\delta_{25}\, \sigma_{th}/RI\right]_f^{25} RI$$

where: $\bar{\sigma}$ = effective microscopic cross section

σ_{th} = thermal microscopic cross section

σ_{25} = measured ratio of epithermal to thermal fissions

RI = infinite dilution resonance integral

Using tabulated values for σ_{th}(19) and RI(20) and a value of δ_{25} = 0.144, a typical value for the lattice in question, we obtain σ_f^{49} = 826 barns and σ_f^{25} = 660 barns. Using these values for σ_f^{49} and σ_f^{25}, a typical value for ICR of 0.5, and the values of N_o^{25} and N^{25} from Table 1 we obtain from Eq. (14) a value of F^{49}/F^{25} = .392.

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